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(54) PROCESS FOR MOLDING MULTI-LAYER ARTICLES

(71) We, ILIKON CORPORATION, a corporation organised and existing under the laws of the State of Delaware, United States of America, of Natick Industrial Centre, Natick, Massachusetts 01760, United States of America, do hereby declare the invention for which we pray that a patent may be granted to us and the method by which it is to be performed to be particularly described in and by the following statement:—

This invention relates to a process for molding multi-layer articles from thermoplastic materials.

At the present time, the use of thermoplastic materials for forming containers for food, particularly beverages, has been severely limited. A primary reason for this is that thermoplastic materials capable of providing an adequate seal between the food and atmosphere are relatively expensive to a degree which has not permitted their economic use in present container forming process at a thickness required to attain the needed structural strength characteristics for the container.

A typical example is carbonated beverages. Since most plastics are poor barriers to carbon dioxide diffusion, only specially formulated polymers can be used, and these are inherently too expensive to be used except for very special purposes. However, a two-layer wall, consisting of a thin, inner layer of the barrier material, backed by a thicker layer of inexpensive material, such as polystyrene or polypropylene, is satisfactory. The raw materials cost will make such a container competitive with glass and considerably less expensive than a bottle made entirely from a barrier material.

Another example of the usage of the multilayer principle would be the combination of a material having good barrier properties for water vapour diffusion with a material having low oxygen permeability. If there is little difference in cost between

the two materials, the layers could be of equal thickness, or in a ratio indicated by ease of manufacturing. It may also, in some cases, be advantageous to have a thin barrier layer on the outside rather than on the inside of a two-layer container, provided the content is compatible with the inner (thick in this case) layer. Another application is where an outside and an inside film encloses a middle layer of recycled, inexpensive plastics, to bury it and to gain from its low cost.

These are but some of many cases where two or more materials together can perform a function that no single material could. It is, therefore, obviously desirable to develop a method for the production of plastics bottles and containers having a composite wall, consisting of two or more layers of different materials. Physical or chemical bonding between the layers is generally unnecessary and may be considered undesirable.

It is an aim of the present invention to provide a process for forming multi-layer containers wherein an adequate barrier can be attained.

According to the present invention there is provided a process for molding multi-layer plastics articles formed from at least two thermoplastics materials which comprises the steps of:

- (a) providing a first injection mold about a mandrel;
- (b) thereafter injecting a first heat-plasticized thermoplastic material into said mold and onto said mandrel to form a first parison;
- (c) thereafter withdrawing said mandrel and first parison from said first injection mold after the parison is formed;
- (d) thereafter cooling at least a portion of said parison on which a second layer is to be injected, to a temperature below the normal working range (as herein de-

fined) of said first thermoplastic material in order to prevent erosion of the first parison by subsequent injection of a second layer thereon;

5 (e) thereafter providing a second injection mold about said mandrel and first parison;

10 (f) thereafter injecting a second heat-plasticized thermoplastic material onto said first parison to form a second layer parison;

(g) thereafter transferring said second layer parison on said mandrel to a blow mold;

15 (h) thereafter blow molding said second layer parison in said blow mold to form an article, and

(i) thereafter removing said article from said mandrel.

20 Between the steps of injecting one layer on top of another layer, it is desirable to provide a conditioning stage, in order to cool at least a portion of the lower layer parison to sufficient rigidity. In this manner, when the next layer is injected on top of a cooled portion of the lower layer, the stream of hot plastics material under high pressure will not erode the lower layer as it might if the lower layer was still relatively hot and thus not sufficiently rigid. Further, such cooling of the lower layer may prevent chemical bonding between layers which bonding is often considered undesirable.

35 If the available heat content of the top layer is sufficiently high, the cooled lower layer may be reheated by the top layer to a temperature sufficient for the blow molding operation. On the other hand, a heating step may be necessary to heat the cooled lower layer to its working temperature.

40 Embodiments of the invention will now be described by way of example with reference to the accompanying drawings in which:

45 Figure 1 is a top sectional view of an apparatus for injection molding two thermoplastics materials followed by blow molding;

50 Figure 2 is a vertical cross-section view of a mold when it is closed around a mandrel, taken along the line 2-2 of Figure 1; and

55 Figure 3 is a top sectional view of a second embodiment of an apparatus for injection blow molding two thermoplastics materials followed by blow molding.

60 As shown in Figure 1, the molding apparatus comprises a turntable 1 having on each side of its periphery a plurality of mandrels 2 shaped to form a parison 3 and a multi-layer parison 4. While a plurality of mandrels are shown, a single mandrel could, of course, be used.

65 In the first step, the thermoplastic mate-

rial is heated in heater 6 to its working temperature and is injected into the manifold 7 and injection nozzles 8. The plastics material is introduced into the space between the molds 9 and the mandrels 2 to form the parisons 3. Each of the molds 9 is formed from two mating mold sections, each mold section being provided with conduits 10 and 11 for passing a fluid there-through thereby maintaining the temperature of the parison as desired.

After the parison 3 is formed, the mold sections are removed from contact with it. The parison 3 is then cooled, for example, by air cooling, to a temperature below its normal working temperature range, i.e. below the temperature at which the plastics material flows. The subsequent injection of a second layer of plastics material, onto the parison 3, at a relatively high flow temperature will not cause erosion of the parison 3. The turntable 1 is then turned through an appropriate angle, which is 90° in the apparatus of Figure 1, so that the parisons 3 are contiguous with the injection nozzles 15. The injection nozzles 15 are associated with the manifold 16 and a heater 17 as well as conduits 10 and 11 in the manner described above for the first injection molding step. A second plastics material is injected over the parison 3 in the second injection molding step. The multi-layer parison 4 is formed from two thermoplastic materials which need not be adherent but may have a similar working temperature and preferably should not react chemically with each other at their molding temperature.

After the multi-layer parison is formed, the turntable 1 is then turned through an appropriate angle so that the multi-layer parisons 4 are contiguous with the corresponding blow molds 20. When the parisons 4 have arrived at the blow molding step, fluid under pressure is introduced through the centre of the mandrel 2 through conduit 22 to expand the parison 4 to the desired final form 23.

After the expansion has been completed, the blow molds 20 are separated into two sections in the same manner as the injection molds thereby exposing the final form multi-layer product to the atmosphere. The turntable 1 is then again turned through an appropriate angle to pick-off station 25. At the pick-off station 25, the final articles have solidified so that they are no longer expandable under fluid pressure. A fluid is then introduced through the conduit 22 through the center portion of the mandrels 2 into the final solidified article 26 to be blown from the mandrel into a collection station, now shown. The turntable 1 is then again turned so that the mandrels 2 are again presented to the injection nozzle 8 to repeat the cycle as described. While we

have described the articles as being blown from the mandrels 2, it will be, of course, understood that other methods of removal may be used.

5 Referring to Figure 2, the molds employed at each injection molding step have interior configurations of slightly increased dimensions while the mold in the blow-molding step has the shape of the final
10 article desired. However, in each step, the mold comprises a top mold section 30 and a bottom mold section 31 attached respectively to platens 32 and 33. In either the blow molding or injection molding steps,
15 the mandrel 2 is brought into position when the mold sections 30 and 31 are apart. After the mandrel has been positioned, the mold sections 30 and 31 are brought together by pressure exerted on platens 32 and 33 so
20 that the mandrel 2 is enclosed by the mold. When the mold sections 30 and 31 are closed, the conduits 10, 11, and a further conduit 35 located in the mandrel are aligned with a source of heating or cooling
25 fluid, not shown, so that during either the blow molding or injection molding steps fluid is passed through these conduits either to heat or cool the parison of the final article. In the blow-molding step, after the
30 article has been expanded, it is sufficiently cooled so that when transferred to the pick-off station it remains intact.

When a final article with more than two layers is desired, the number of injection
35 molding steps is increased. Thus, for forming a three layer article, a 5 sided turntable is used, and the turntable is turned 72° between each step of the process. When a 4-layer article is desired, the turntable will
40 have six sides, and the table is turned 60° between each step of the process, and similarly for fabricating articles with more than 4 layers.

Particularly suitable thermoplastic materials for use with the present invention
45 include polyethylene with Saran Registered Trade Mark (polyvinylidene chloride), acrylics with acrylonitrile-ethylacrylate copolymer (2.g., Barex Registered Trade Mark
50 210), polystyrene with Barex 210, polypropylene with Barex Registered Trade Mark 210, or acellulosic such as ethyl cellulose, cellulose acetate, cellulose acetate-butyrate
55 cellulose propionate or the like with Barex 210.

Referring to Figure 3, a seven-sided turntable 40 is shown therein having on each side of its periphery two mandrels 2' shaped to form a parison 3' and a multi-layered
60 parison 4'. The particular number of mandrels carried by each side of the turntable is merely a matter of choice or design.

In the first injection station 38, thermoplastic material is heated in heaters 6' to
65 its working temperature and is injected into

the manifold 7' and injection nozzles 8'. The plastic is introduced into the space between the molds 9' and the mandrels 2' to form the parisons 3'. Each of the molds 9' is formed from two mating mold sections,
70 each mold section being provided with cooling channels for the passing of fluid there-through for maintaining the temperature of the parison as desired.

After the parison 3' is formed, the mold
75 sections are removed from contact with it. The turntable 40 is then turned through an appropriate angle, which is $51\frac{3}{7}^\circ$ in the apparatus of Figure 3, to a conditioning
80 station 42. The conditioning station has been found to be desirable because when the second layer is injected on top of parison 3' the stream of hot plastic under high pressure may erode parison 3' if parison 3'
85 has not obtained sufficient heat loss by cooling. Thus an intermediate conditioning station 42 is provided between the first and second injection stations in order to cool, to sufficient rigidity, at least the portion of the first layer parison on which the second
90 layer is to be injected.

Conditioning station 42 may comprise a chill plate for cooling at least a portion of the first layer parison 3'. The cooling in certain instances could be effected by circulating cooling fluid through the mandrel.
95 In another method, the parison could be refrigerated i.e. placed in a cold or cool atmosphere or else air cooling can be utilized where sufficient, without requiring a separate conditioning station.

By using a cooling step, the various layers of plastics material need not be workable in the same temperature range. For example, if the inner layer of material requires a lower injection temperature than the second layer, the inner layer parison 3' can be cooled to a temperature below its normal working range. Heat from the second layer will then diffuse into this cool
105 layer and reheat it. By correctly designing the variables, such as degree of cooling, temperature and size of the second layer injection and cycling time, the first layer can be brought back to its proper working temperature without sufficiently cooling the second layer down below its working range.

Thus, referring to Figure 3 after parison 3' has been cooled in conditioning station 42, the turntable 40 is turned to the second
120 injection station 44. At injection station 44, thermoplastic material is heated in heaters 17' to its working temperature and is injected into the heater nozzles 16' and injection nozzles 15' so that the second plastics
125 material will be injected over the parison 3' in the second molding step. The multi-layer parison 4' is formed from the two thermoplastic materials.

If the available heat content in the sec- 130

ond layer is sufficiently high, the relatively cool first layer will be reheated by the second layer to a temperature adequate for the blow molding operation. On the other hand, if the available heat content in the second layer is not sufficiently high, another conditioning station 48 will be needed between the second injection station 44 and the blow molding station 46, in order to re-introduce heat into the first layer.

It can be seen that the conditioning station 48 is provided between second injection station 44 and blow molding station 46. The second conditioning station 48 is utilized to reintroduce heat into the first layer when necessary and such reintroduction of heat may be obtained either by circulating a hot fluid through internal channels in the mandrel, by high frequency induction heating of the mandrel using an external coil, or by dielectric heating of the first layer using a high frequency which will generate heat mostly in the first layer. The dielectric heating method will only be possible if the dielectric characteristics of the layers are different enough on some frequency range to enable selective heating.

As stated above, certain materials will not require conditioning station 48 because heat from the second layer will be sufficient to reheat the first layer. However, if the working temperature of the first layer material is higher than the working temperature of the second layer materials, it is not possible to reheat the first layer by the latent heat in the second layer injection. Likewise, it is not possible to reheat the first layer if other processing parameters do not allow sufficient time for proper heat transfer, even if enough latent heat is available. It is, therefore, necessary to reheat the first layer parison 3' in conditioning station 48 in one of the manners described above. Although production of a two layered article is shown in Figure 3, it is to be understood that the present invention is also useful with the manufacture of three or more layered articles.

Referring back to Figure 3, after the two layered parison 4' is conditioned for further working in conditioning station 48, turntable 40 is then turned so that the two layer parisons 4' are contiguous with the corresponding blow molds 20'. When the parisons 4' have arrived at the blow molding step, fluid under pressure is introduced through the centre of the mandrel 2' to expand the parison 4' to the desired final form 23'. After the expansion has been completed, the blow molds 20' are separated into two sections in the same manner as the injection molds thereby exposing the final form multi-layer product to the atmosphere. The turntable 40 is then again turned through an appropriate angle to ejection station 50.

At ejection station 50, the final articles have solidified so that they are no longer expandable under fluid pressure. Fluid is then introduced through the centre portion of the mandrels 2' into the final article, to eject it from the mandrel into a collection station.

It may be necessary in some cases to provide a conditioning station 52 between the ejection station 50 and the first injection station 38, in order to bring the mandrel temperature to the proper level for the first stage injection.

In the production of bottles, the mandrels utilized are relatively long and slender and since it is virtually impossible to obtain absolutely even flow of plastics into the mold cavity during the first injection station, the mandrel will be subjected to elastic bending. Such bending is of little consequence in the production of typical one layer bottles, since the amount of bending represents only a small fraction of the parison wall. On the other hand, with a relatively thinwall parison, the bending is substantial enough to cause marked eccentricity of the parison. In the blowing stage, this could lead to uneven wall thickness or may result in rupture of the inner layer.

In order to alleviate the eccentricity problem it is preferred to utilize a mandrel that is formed from a material with a high elastic modulus. One possibility is tungsten which has an elastic modulus of 51.6×10^6 pounds per square inch and another material which could be used is cobalt-infiltrated tungsten which has an elastic modulus of between 70×10^6 pounds per square inch and 90×10^6 pounds per square inch. As a comparison, conventional mandrels are formed of steel having an elastic modulus of about 30×10^6 pounds per square inch. Since bending at a given stress is inversely proportional to the modulus, it is thus possible to reduce the bending of the mandrel significantly by utilizing a mandrel formed from a material with a very high elastic modulus.

Another system for alleviating the eccentricity problem is by securing the tip of the mandrel relative to the mold during the initial portion of the injection physically to maintain the mandrel properly centered in the mold cavity. The thermoplastic material would be injected from the neck of the mandrel, instead of from its tip and as injection progresses the device is withdrawn to enable plastics material to flow freely in its place.

WHAT WE CLAIM IS:—

1. A process for molding multi-layer plastics articles formed from at least two thermoplastic materials which comprises the steps of:

(a) providing a first injection mold about

a mandrel,

(b) thereafter injecting a first heat-plasticized thermoplastic material into said mold and onto said mandrel to form a first parison,

(c) thereafter withdrawing said mandrel and first parison from said first injection mold after the parison is formed,

(d) thereafter cooling at least a portion of said parison on which a second layer is to be injected, to a temperature below the normal working range as herein defined of said first thermoplastic material in order to prevent erosion of the first parison by subsequent injection of a second layer thereon,

(e) thereafter providing a second injection mold about said mandrel and first parison,

(f) thereafter injecting a second heat-plasticized thermoplastic material onto said first parison to form a second layer parison,

(g) thereafter transferring said second layer parison on said mandrel to a blow mold,

(h) thereafter blow molding said second layer parison in said blow mold to form an article, and

(i) thereafter removing said article from said mandrel.

2. A process according to Claim 1, including the step, after the second layer parison is formed on said mandrel, of withdrawing said mandrel and second layer parison from said second injection mold and thereafter heating said second layer parison so that said first thermoplastic material and said second thermoplastic material are both at a temperature that is high enough for subsequent blow molding thereof.

3. A process for molding multi-layer plastics articles formed from at least two thermoplastic materials which comprises the steps of: injecting a first heat-plasticized thermoplastic material onto a mandrel in an injection mold to form a parison; removing said injection mold after the parison is formed; thereafter cooling at least a portion of said parison on which another layer is

to be injected, to a temperature below the normal working range as herein defined of said first thermoplastic material in order to prevent erosion of the first parison by subsequent injection of another parison layer thereon, transferring said parison to at least one additional injection mold, injecting a second heat-plasticized thermoplastic material onto said parison at each additional injection molding step to form a multi-layer parison; after each parison is formed removing the injection mold and cooling at least a portion of the parison formed between each injection step so that at least a portion of the top layer, prior to injection of the next injection layer, is at a temperature below the normal working range as herein defined of the thermoplastic material from which the parison is formed in order to prevent erosion of the formed parison by subsequent injection of another parison layer thereon; transferring said multi-layer parison on said mandrel to a blow mold; blow molding said multi-layer parison in said blow mold to form an article; and removing said article from said mandrel.

4. A process according to Claim 1, wherein said first layer parison is cooled by a chill plate.

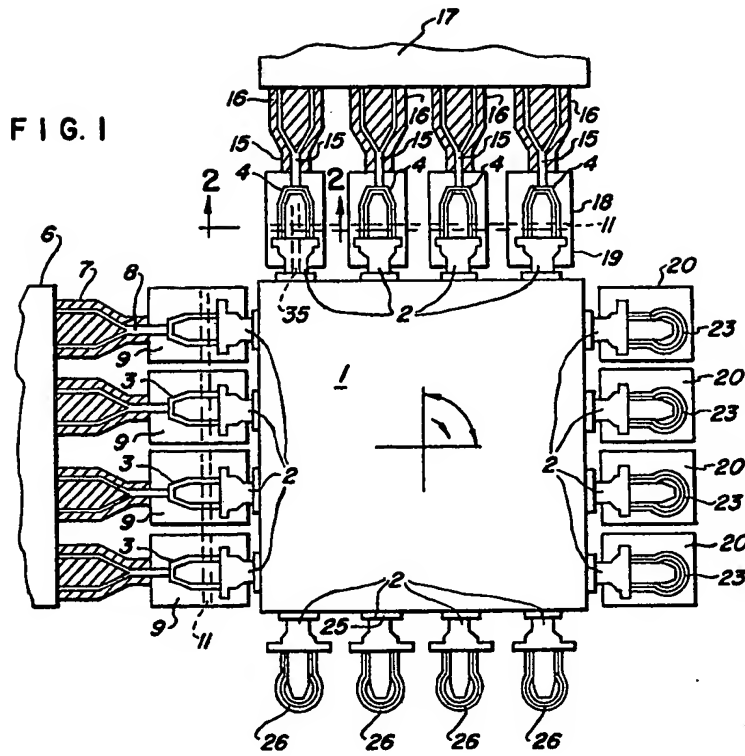
5. A process according to Claim 1, wherein said first layer parison is cooled by circulating fluid through said mandrel.

6. A process according to Claim 1, wherein said first layer parison is cooled by refrigeration in a cold or cool atmosphere.

7. A process according to Claim 1, wherein said first layer parison is air cooled.

8. A process for molding multi-layer plastics articles formed from at least two thermoplastic materials, substantially as herein described with reference to Figures 1 and 2 or Figure 3 of the accompanying drawings.

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**FIG. 2**